

Critical Properties of Thin Liquid Layers¹

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ABSTRACT

The physical properties of thin liquid layers are studied. The notion of spatial limitation of a system undergoing critical phenomena and phase transitions is proposed. The scaling formulae for the singular part of the fluctuation free energy and correlation length for liquid systems are obtained and analyzed. The method to study the pair correlation function and correlation length of order parameter fluctuations for thin liquid layers is proposed. The connection of results obtained for thin liquid layers with the Fisher scaling hypothesis for finite-size systems is discussed.

KEY WORDS: correlation length, critical state, finite-size system, scaling hypothesis, statistical mechanics, thin liquid layers.

1. INTRODUCTION

The physical properties of systems undergoing critical phenomena and phase transitions are essentially influenced by such a practical factor as the spatial limitation. Many systems of experimental and theoretical interest are spatially finite-sized, such as thin liquid layers, interfaces, porous media, biomembranes, synaptic clefts, etc.

Here we proposed an approach to studying critical phenomena in thin liquid layers based on the methods of statistical physics, namely the integral and corresponding differential equations for the pair correlation function of order parameter fluctuations (see e.g. [1-5]) and on the scaling hypothesis for finite-size systems proposed by M.Fisher [6]. The main idea of our approach is connected with the fact that the pair correlation function can be found as the Green function for the Helmholtz operator with the certain boundary conditions at the surfaces of a thin plane-parallel layer. It allows then to study the critical behavior of thin liquid layers and discuss the physical properties of experimental interest.

2. THE NOTION OF SPATIAL LIMITATION AND GENERAL PROPERTIES OF FINITE-SIZE LIQUID SYSTEMS

2.1. The notion of spatial limitation

A system can be considered as finite-sized near the critical point the phase transition point if its characteristic linear size L becomes comparable with the correlation length R_c of the order parameter fluctuation, i.e. $L_i \approx R_c$. As is well known from the fluctuation (scaling) theory of phase transitions (see e.g. [7]), the correlation length $R_c = R_{co}\tau^v$, where the amplitude of the correlation length in liquids can reach the value $R_{co} \approx 1-10$ nm. Nowadays, it became possible to reach the critical point with

respect to the temperature variable at “distances” like $|T - T_c|_{\min} \approx 10^{-2} - 10^{-3}$ K for $\tau \approx 10^2$, or using dimensionless values $\tau_{\min} = |T - T_c| / T_c \approx 10^{-5}$ [7-10]. As the critical exponent of the correlation length is equal $\nu \approx 0,63$ for the space dimensionality $d = 3$, then the maximum values of the correlation length in classical liquids can approach such values as $R_c \approx 1 - 10 \text{ nm} \cdot (10^{-5})^{-0,6} \approx 10^3 - 10^4 \text{ nm}$. (Spatial situation can be realized for quantum liquids where, because of very small values of the critical temperature, the correlation length becomes two orders more; for instance, in liquid helium $\tau \approx 10^{-8}$ $R_c / R_{co} \approx 10^5$). So, liquid systems which have the linear size L about 1 - 10 mkm could be considered as finite-sized near the critical point.

2.2. General properties of finite-size liquid systems

M.Fisher [6] and other investigators (see e.g. [11,12]) have proposed a modified scaling hypothesis for such finite-size systems. According to this hypothesis (here we shall formulate it for classical liquids), the fluctuation part of the thermodynamic potential and the correlation length depend not only on the thermodynamic variables (the reduced temperature τ , etc.) and external fields h but on the linear size L of a system:

$$\Delta \dots = L^{-d} f(a\tau L^{1/\nu}, bhL^{\beta\delta/\nu}),$$

$$R_c = L f_R(a\tau L^{1/\nu}, bhL^{\beta\delta/\nu}). \quad (1)$$

The first scaling argument in Eq. (1) $= a\tau L^{1/\nu}$ in both scaling functions f and f_R can be obtained from the following formulae:

$$R_c \sim \tau^{-\nu}, \quad \tau \sim R_c^{-1/\nu}, \quad x \sim \frac{\tau}{R_c^{-1/\nu}} \sim \tau L^{1/\nu} \quad (R_c \rightarrow L) \quad (2)$$

In analogous way it is easily to obtain the expression for the second scaling argument $= bhL^{\beta\delta/\nu}$ with taking such formulae into account:

$$R_c \sim \tau^{-\nu} \sim \varphi^{-\nu/\beta} \sim h^{-\nu/\beta\delta}, \quad \varphi \sim \tau^\beta, \quad \varphi \sim h^{1/\delta}, \quad \tau \sim \varphi^{1/\beta},$$

$$h \sim R_c^{-\beta\delta/\nu}, \quad y \sim \frac{h}{R_c^{-\beta\delta/\nu}} \sim hL^{\beta\delta/\nu} \quad (R_c \rightarrow L) \quad (3)$$

Here $\varphi = (\rho - \rho_c) / \rho_c$ is the order parameter for classical liquids, i.e. the reduced density.

Let us consider some consequences of the scaling hypothesis (1) for the physical properties of finite-size liquids.

) equilibrium value of the order parameter (the reduced density or concentration)

$$\varphi_0 = \{ \rho_0 - \rho_c, \dots \} = (\partial \Delta / \partial h)_\tau = L^{-d+\beta\delta/\nu} f_\varphi(x, y),$$

(4)

$$f_\varphi = b(f_\varphi)'_h, \quad -d + \beta\delta/\nu = -\beta/\nu \quad (3\nu = 2 - \alpha, \alpha + 2\beta + \gamma = 2, \beta\delta = \beta + \gamma).$$

Thus, it is possible to propose such an algorithm to derive the dependence of any physical property on the linear size L in a finite-size system:

$$\sim \tau^{-n} \sim R_c^{n/\nu} \sim L^{n/\nu}. \quad (5)$$

In other words, if any quantity has its singular behavior $\sim \tau^{-n}$ in the spatially infinite system, then with taking such formulae into account

$$R_c \sim \tau^{-\nu}, \quad \tau \sim R_c^{-1/\nu}, \quad R_c \rightarrow L$$

the dependence of this quantity on the linear size L in a finite-size system near its critical point or phase transition point must be as follows:

$$\sim L^{n/\nu}. \quad (6)$$

For instance, for the order parameter in the spatially infinite system one has $\varphi_0 \sim \tau^\beta$.

Then in a finite-size system the dependence of the order parameter on the linear size L is

$\varphi_0 \sim \tau^\beta \sim R_c^{-\beta/\nu} \sim L^{-\beta/\nu}$. It gives directly such a result for a spatially infinite system where the linear size and correlation length are going to infinity ($L, R_c \rightarrow \infty$): the value of the order parameter is vanishing ($\varphi_0 \rightarrow 0$) while approaching the critical state, as it is expected for this system.

b) the isothermal compressibility of finite-size liquids

$$\beta_T = (\partial^2 \Delta / \partial h^2)_\tau = L^{-d+2\beta\delta/\nu} f_\beta(x, y), \quad ,$$

$$f_\beta = b(f_\varphi)''_h, \quad -d + 2\beta\delta/\nu = \gamma/\nu. \quad (7)$$

c) the heat capacity of finite-size liquids

$$= (\partial^2 \Delta / \partial \tau^2)_h = L^{-d+2/\nu} f_c(x, y), \quad (8)$$

$$f_c = a(f_\varphi)''_\tau, \quad -d + 2/\nu = \alpha/\nu.$$

d) shifts of the critical parameters in finite-size liquids (the critical temperature, density, concentration, critical external fields). It is possible to show that the critical parameters for any finite-size liquid are not unique. Say, the critical temperature of a certain spatially limited liquid varies for various physical properties.

Let us find a new value of the critical temperature $T_c(L)$ in compare with its bulk value $T_c(\infty)$ for a spatially infinite liquid. First, we shall find the shift of the critical temperature $\tau^* = [T_c(L) - T_c(\infty)] / T_c(\infty)$ for the isothermal compressibility or for the susceptibility in a general case. One has the following expression for this quantity:

$$\chi = L^{\gamma/\nu} f_\chi(x, y) \underset{x=a\tau^{1/\nu}}{=} \left(\frac{x}{a\tau} \right)^\gamma f_\chi(x, y)$$

and the corresponding condition of its maximum

$$\left(\frac{\partial \chi}{\partial \tau} \right)_h = -\gamma \left(\frac{x}{a\tau^*} \right)^\gamma \frac{1}{\tau^*} f_\chi + \left(\frac{x}{a\tau^*} \right)^\gamma f'_\chi aL^{1/\nu} = 0. \quad (9)$$

It is easy to obtain from Eq. (9) such a formula for the shift of the critical temperature

$$\tau^* = \frac{T_c(L) - T_c(\infty)}{T_c(\infty)} = \frac{\gamma f_\chi}{a f'_\chi} L^{-1/\nu} = A_\chi L^{-1/\nu}. \quad (10)$$

Now let us find the shift of the critical temperature for the heat capacity. It gives such results

$$\begin{aligned} &= L^{\alpha/\nu} f_c(x, y) = (x/a\tau)^\alpha f_c(a\tau L^{1/\nu}, y) \\ &-\alpha \left(\frac{x}{a\tau^*} \right)^\alpha \frac{1}{\tau^*} f_c + \left(\frac{x}{a\tau^*} \right)^\alpha f'_c a L^{1/\nu} = 0, \\ &\tau^* = \frac{\alpha f_c}{a f'_c} L^{-1/\nu} = A_c L^{-1/\nu}. \end{aligned} \quad (11)$$

Finally one has such a formula for the shift of the critical temperature from the condition of maximum for the correlation length R_c :

$$\tau^* = \frac{\nu f_R}{a f'_R} L^{-1/\nu} = A_R L^{-1/\nu}. \quad (12)$$

Thus, it is possible to make the important conclusions as follows:

1. Eqs. (10)-(12) give the *universal* behavior of the size dependence $\tau^* \sim L^{-1/\nu}$ for shifts of the critical temperature in finite-size systems.
2. Numerical values of τ^* which give maximums for various physical properties in finite-size systems (χ , R_c , ...) are *not universal* because of the different amplitudes $\chi \neq R_c$ in Eqs. (10)-(12).

Experimental studies by H.Lutz et al. [13] and others (see e.g. [14]) confirm the size dependence $\tau^* \sim [T_c(L) - T_c(\infty)] \sim L^{-1/\nu}$ for shifts of the critical temperature in finite-size systems.

Summarizing the critical behavior of finite-size systems in compare with infinite ones, it is worthy to stress the most principal results, namely:

- existence of the above-mentioned shifts of the critical temperature and other critical parameters,
- existence of the new values of the critical exponents,
- existence of the certain vicinity of phase transition which becomes narrow while the linear size of a system is increasing,
- existence of the shifts of maximums for physical properties at the new critical temperature (other critical parameters) towards large temperatures (densities, etc.) and an anomalous growth of these maximums (without taking the effects of spatial dispersion into account) with transition to spatially infinite systems.

3. PAIR CORRELATION FUNCTION AND CORRELATION LENGTH OF THIN LIQUID LAYERS

3.1. Pair correlation function

An anomalous growth of the correlation length R_c predicted by the scaling-law theory [7] can realize only for spatially infinite systems. Only for such systems the following expression for the pair correlation function from the Ornstein-Zernicke (OZ) approximation is valid:

$$G_2(r) = A \exp (-r/R_c)/r, \quad (13)$$

The pair correlation function of order parameter fluctuations in Eq. (13) is the Green $\hat{L} = \nabla^2 - R_c^{-2}$ for a spatially infinite system with zeroth boundary conditions when the distance r between two fluctuation is going to infinity. To use Eq. (13) for systems of

experimental interest, it is necessary for linear size of a system L to be much more than the correlation length R_c .

Let us assume the geometry of our thin liquid system in the form of a plane-parallel layer $-\infty < x, y < \infty, -L_0 \leq z \leq L_0$. One can find the pair correlation function Green function $G_2(R)$ as the Green function of the differential Helmholtz operator for such a thin liquid layer with zeroth boundary conditions at the surfaces $z=L_0$ and $z=-L_0$.

There is another method [4,5] of finding the pair correlation function as the result of acting of the inverse Helmholtz operator $\hat{L} = \nabla^2 - \kappa^2$ (here $\kappa = R_c^{-1}$) on the delta-function $\delta(r)$. To solve this complicated problem, it first requires the formula for the delta-function $\delta(r)$ constructed of orthonormal eigenfunctions for the Helmholtz operator \hat{L} with zeroth boundary conditions for this function together with its first derivative and with eigenvalues satisfying the relation $\lambda_n = k_x^2 + k_y^2 + k_z^2 + \kappa^2$. Here components of wave vector k_x, k_y change continuously from $-\infty$ to $+\infty$, while k_z is discrete ($k_z = n^2 \pi^2 / 4L_0^2, n = 0, 1, \dots$). The following formula for the delta-function satisfied all these conditions:

$$\delta(r) = (1/8\pi L_0) \sum_{n \geq 0} \iint [1 - (-1)^n] \cos(\pi n z / 2L_0) e^{i(k_x x + k_y y)} dk_x dk_y \quad (14)$$

With allowance for Eq. (14), one can obtain an expression for the pair correlation function of order parameter fluctuations in the form

$$G_2(r) = \hat{L}^{-1} \delta(r) = (1/8\pi^2 L_0^2) \sum_{n \geq 0} \iint [1 - (-1)^n] \cos(\pi n z / 2L_0) e^{i(k_x x + k_y y)} (k_x^2 + k_y^2 + (n\pi/2L_0)^2 + \kappa^2)^{-1} dk_x dk_y \quad (15)$$

Then it is convenient to use polar coordinates

$$k_x = k_{xy} \cos \varphi, \quad k_y = k_{xy} \sin \varphi, \quad \rho = (x^2 + y^2)^{1/2}$$

and to fulfil the integration in (15) with taking such formulae for cylindrical functions into account

$$(ik\rho\cos\varphi) = J_0(k\rho) + 2i\cos\varphi J_1(k\rho) + 2i^2\cos 2\varphi J_2(k\rho) + \dots,$$

$$\int_0^\infty \frac{k J_0(k\rho) dk}{k^2 + a^2} = K_0(\rho a).$$

It gives the following expression for the pair correlation function $G_2(r, z)$ of a thin liquid layer:

$$G_2(p, z) = (1/4\pi L_0) \sum_{n \geq 0} [1 - (-1)^n] K_0 \left[\rho \left(\hat{e}^2 + \frac{n^2 \pi^2}{4L_0^2} \right)^{1/2} \right] \cos(n\pi z / 2L_0). \quad (16)$$

Here $K_0(u)$ is the cylindrical Macdonald function.

In the case of a plane-parallel layer of thickness $d = 2L_0 \gg R_c$ the limiting transition to the OZ approximation can be easily obtained:

$$\begin{aligned} G_2(p, z) &= (1/4\pi L_0)(2L_0/\pi)^2 \int_0^\infty K_0 \left[\rho \left(\hat{e}^2 + \frac{n^2 \pi^2}{4L_0^2} \right)^{1/2} \right] \cos(k_z z) dk_z = \\ &= (1/4\pi) \exp[-(\rho^2 + z^2)^{1/2}] / (\rho^2 + z^2)^{1/2}. \end{aligned} \quad (17)$$

The main contribution in Eq. (17) for $G_2(p, z)$ is given by the first term. Therefore, it is possible to use an approximate expression for the pair correlation function of density fluctuations in thin liquid layer

$$G_2(p, z) = (1/2\pi L_0) K_0 \left[\rho \left(\hat{e}^2 + \pi^2 / 4L_0^2 \right)^{1/2} \right] \cos(\pi z / 2L_0). \quad (18)$$

In accordance with Eqs. (16) and (18), the correlation function $G_2(r)$ demonstrates an oscillatory behavior in the ρ direction confirming computer-simulation studies for the radial distribution function in finite-size liquids.

3.2. Correlation length of density fluctuations

Because of a non-exponential shape of G_2 , it is natural to find the correlation length of density fluctuation as the second normalized spatial moment of the pair correlation function

$$R_c^* = (\langle r^2 \rangle)^{1/2}, \quad \langle r^2 \rangle = \int r^2 G_2(r) dr / \int G_2(r) dr. \quad (19)$$

Using polar coordinates in Eq. (19) and formula for the main contribution for the pair correlation function given by Eq. (18), one has

$$\langle r^2 \rangle = \frac{\int_0^\infty \rho^3 K_0(\rho \sqrt{\hat{\epsilon}^2 + \pi^2 / 4L_0^2}) d\rho}{\int_0^\infty \rho^3 K_0(\rho \sqrt{\hat{\epsilon}^2 + \pi^2 / 4L_0^2}) d\rho} \quad (20)$$

With taking the following expression for the cylindrical functions into account

$$\int_0^\infty x^\mu K_\nu(ax) dx = 2^{\mu-1} a^{\mu-1} \tilde{A} \left(\frac{1+\mu+\nu}{2} \right) \tilde{B} \left(\frac{1+\mu-\nu}{2} \right),$$

it is possible to derive the formula for the correlation length of density fluctuations:

$$R_c = [\frac{4}{\hat{\epsilon}^2 + \pi^2 / 4L_0^2} + (1 - 8/\pi^2) 4L_0^2]^{1/2}. \quad (21)$$

The consequences can be deduced from Eq. (21) as follows:

1. The correlation length R_c of thin liquid layer depends not only upon thermodynamic variables (because of relationships between κ and temperature, density, etc.) but also on the thickness of a layer $d=2L_0$ or on the geometric factor $K=d/R_{c0}$ which shows the number of molecular monolayers along the direction of spatial limitation.
2. The correlation length expressed by Eq. (21) is determined by two contributions

$$R_c = [(R_c)_{xy}^2 + (R_c)_z^2]^{1/2}$$

where

$$(R_c)_{xy} = \frac{2}{(\hat{\epsilon}^2 + \pi^2 / 4L_0^2)^{1/2}}$$

is the correlation length in the xy plane, and

$$(R_c)_z = 2L_0(1 - 8/\pi^2)^{1/2}$$

is the correlation length in the z direction.

3. At the critical bulk point the value χ^{-2} (the inverse isothermal compressibility) is going to zero and one has for the correlation length in the xy plane such an expression

$$(R_c)_{xy} = 4L_0 / \pi.$$

If the thickness of a plane-parallel liquid layer is equal $d=2L_0 \approx 100$ nm, then

$$(R_c)_{xy} = 63,7 \text{ nm at } T=T_c(\infty).$$

4. For the case of the critical density and $\rho \neq \rho_c$ the value $\chi^{-2} \sim (\partial \mu / \partial \rho) \sim \tau^\gamma$,

or $\chi^{-2} = \chi_0^{-2} \tau^{5/4}$, where χ_0 is the inverse amplitude of the correlation length ($\chi_0 = R_{co}^{-1}$). The amplitude of the correlation length R_{co} has usually the same order of magnitude as the radius a_0 of direct intermolecular interactions, i.e. $\chi_0 \approx a_0^{-1} \approx (0,1 \text{ nm})^{-1} \approx 10^{10} \text{ m}^{-1}$.

¹. Then if $L_0 = \chi_0^{-1} \chi_0$ (coefficient $\chi_1 = L_0 / a_0 = 500$ for thickness $2L_0 = 10^2$ nm) one has for the correlation length in the xy plane

$$R_c(\tau)_{xy} = \frac{2 \cdot 10^{-10} \hat{\chi}}{\sqrt{\tau^{5/4} + \pi^2 / 4\hat{\epsilon}_1^2}}. \quad (22)$$

Numerical values of $R_c(\tau)_{xy}$ with respect to the reduced temperature τ are illustrated by Fig.1.

5. For the case of $T = T_c$ and $\rho \neq \rho_c$ the value $\chi^{-2} \sim (\partial \mu / \partial \rho) \sim \Delta \rho^{\delta-1}$, where

$\Delta \rho = (\rho - \rho_c) / \rho_c$, or $\chi^{-2} = \chi_0^{-2} \Delta \rho^{3,5}$. For the same parameters χ_0 , R_{co} , L_0

and ρ_c as in the case of the critical density one has the following expression for the correlation length in the xy plane:

$$R_c(\Delta\rho)_{xy} = \frac{2 \cdot 10^{-8} \hat{t}}{\sqrt{|\Delta\rho|^{3.5} + \pi^2 / 4 \hat{e}_1^2}} \quad (23)$$

Fig. 2 illustrates the density dependence of the correlation length of a plane-parallel layer given by Eq. (23).

4. CONNECTION WITH THE SCALING HYPOTHESIS FOR FINITE-SIZE SYSTEMS

The initial scaling hypothesis for finite-size systems has been formulated by M.Fisher in [6]. Let us discuss results obtained above for thin liquid layers with this hypothesis formulated for the susceptibility

$$\chi = K^\omega F(K^\theta \tau) \quad (24)$$

Here K is the above-mentioned geometric factor and $F(x)$ is the corresponding scaling function of the susceptibility (the isothermal compressibility in liquids). It is worthy to mention that Eq. (24) and the scaling hypothesis for finite-size systems formulated in Eqs. (1) and (7) give the same results for the susceptibility and other physical properties in zeroth external field, i.e. for the second scaling argument $y = 0$ in scaling functions of Eqs. (1) and (7).

The most important consequences of Eq. (24) are as follows:

- the critical exponent $\omega = 2 - \eta$;
- the critical exponent $\theta = 1/\nu$;

- the scaling function $F(x)$ has such an asymptotic formula for large arguments $x = K^\theta \tau \gg 1$: $F(x) \sim x^{-\gamma}$, where $\gamma = (2-\eta)\nu$ is the critical exponent of the susceptibility for bulk systems (isothermal compressibility for bulk liquids);
- the scaling function $F(x)$ has such an asymptotic formula for small arguments when $x \rightarrow x_c$: $F(x) \sim (x - x_c)^{-\zeta}$ where ζ is not equal in general case to γ .

Let us verify the validity of the scaling hypothesis (24). With allowance for formulae (18) and (21), one has such an expression for the susceptibility for a thin liquid plane-parallel layer

$$\chi = \chi_0 K^2 \{ 4 [K^2 \tau^{2\nu} f_I(x) + \pi^2]^{-1} + (1 - 8/\pi^2)/4 \} \quad (25)$$

where χ_0 is the amplitude of the susceptibility.

Comparison between Eqs. (24) and (25) gives the following results:

1. According to Eqs. (24) and (25), the critical exponent $\omega = 2$. Taking into account that $\omega = \gamma/\nu = 2 - \eta$ and the fact that the Helmholtz differential operator \hat{L} corresponds to the OZ approximation with zeroth value of the critical exponent η , this result confirms the first consequence of the Fisher scaling hypothesis (24) for finite size systems.
2. Scaling function $F(x)$ in Eq. (25) depends on the argument $x = K^{2/\gamma} \tau$ for zeroth external field, i.e. in the vicinity of the critical isochore. Therefore, for $\eta = 0$ one has $2/\gamma = 1/\nu = \theta$ in accordance with the second consequence of the scaling hypothesis (24).
3. With increasing the thickness of a plane-parallel liquid layer one has the following result for the scaling function $F(x)$ from Eq. (25) for large arguments

$x = K^{2/\gamma} \tau \gg 1$: $F(x) \sim x^{-1} \sim x^{-\gamma}$ in accordance with the third consequence of the scaling hypothesis (24).

4. The asymptotic behavior of the scaling function $F(x)$ from Eq. (25) for small arguments $x = K^{2/\gamma} \tau \ll 1$ does not agree with the last consequence of the scaling hypothesis (24). It could take place only if $\zeta = \gamma$. This result means that we cannot use our approach in presented form to describe the transition from 3-dimensional finite-size liquid layers to 2-dimensional monolayer. The probable reason is connected with the fact that we have found only a nonsingular asymptotic solution for the pair correlation function G_2 which is not valid for very small geometric factor ($K \rightarrow 1$).

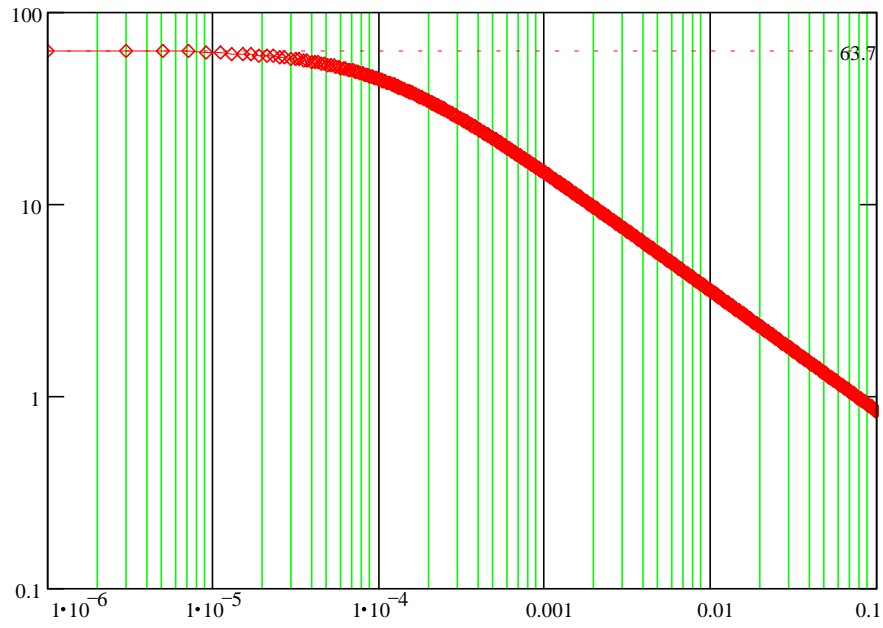
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FIGURE CAPTIONS

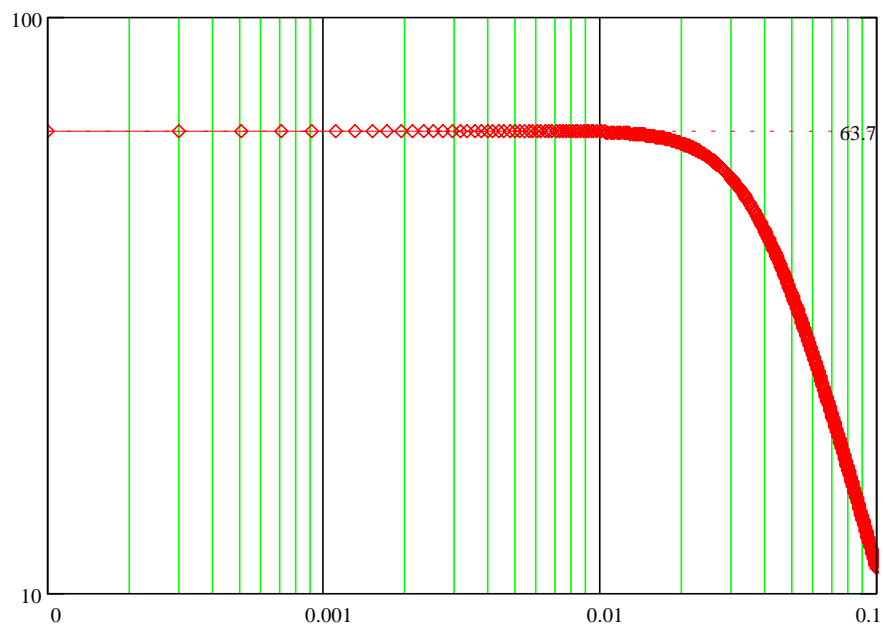
Fig. 1. Temperature dependence of the correlation length $(R_c)_{xy}$, nm for a thin liquid layer ($d=100$ nm, $a_0=R_{co}=0,1$ nm, $k_1=500$, $\gamma=5/4$).

Fig. 2. Density dependence of the correlation length $(R_c)_{xy}$, nm for a thin liquid layer ($d=100$ nm, $a_0=R_{co}=0,1$ nm, $k_1=500$, $\delta=4,5$).



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Fig. 1.



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Fig. 2.